DISPERSION OF X-RAYS IN CALCITE

By Louis A. Pardue
Yale University

(Received September 23, 1931)

Abstract

The dispersion of x-rays in calcite has been investigated. The total radiation and the $K\alpha_1$ line from a molybdenum target tube operated at about 44,000 volts were used in different experiments. The decrement of unity in the index of refraction for the $K\alpha_1$ wave-length was found to be $(2.001 \pm 0.009) \times 10^{-6}$. This does not agree with the value computed on the Drude-Lorentz theory. The specimen was a right prism with the optic axis parallel to the 90° refracting edge. No evidence was found for double refraction. The intensities of Mo$K\alpha_1$ radiation reflected from calcite mirrors were measured for angles in the neighborhood of the critical angle. These experimental values were compared with the values computed on the basis of Thibaud's modification of Fresnel's equation and found to be in fair agreement.

OPTICAL terminology and theory may be applied to x-rays if due account is taken of their higher frequency. Since the different theories of the dispersion of x-rays agree in the limiting case where the frequency of the radiation is much greater than the natural frequency of the electrons in the dispersing medium, the results (Table I) for the refractive index of calcite for Mo$K\alpha_1$ are suggestive. The values reported for δ are considerably larger than the value resulting from all theories in the limiting case for higher frequencies. The theoretical value is

$$\delta \equiv 1 - \mu = ne^2/2\pi m\nu^2$$

where μ is the index of refraction, n the number of electrons per cm^3 in the dispersing medium, e the charge on an electron, m the electronic mass, and ν the frequency of the radiation. Table I gives the results of the various investigators and their methods.

Table I.

	$(1-\mu)\ (10)^6$ Theor.	Exper.	Method
Davis and Hatley[1]	1.84	2.0	Crystal wedge
Hatley[2]	1.84	2.03 ± 0.09	Crystal wedge
Compton[3]	1.84	2.10 ± 0.15	Deviation

[1] C. C. Hatley, B. Davis, Phys. Rev. [2] **23**, 290 (1924).
[2] C. C. Hatley, Phys. Rev. [2] **24**, 486 (1924).
[3] A. H. Compton, Phys. Rev. [2] **37**, 1694 (1931).

While the probable errors are large there is a definite lack of correspondence between theory and experiment. It was deemed desirable to make a very careful study of dispersion of Mo$K\alpha_1$ x-rays in calcite, having in mind the possibility of double refraction, about the only phenomenon in optics

whose counterpart in x-rays has escaped detection. The specimen of calcite chosen was a right prism with the optic axis parallel to the 90° refracting edge. To have the optic axis so directed is a logical choice in seeking double refraction in x-rays but one without strong support. An investigation was made of the energy reflected from polished and cleaved faces of calcite at angles in the neighborhood of the critical angle.

Arrangement and Adjustment of Apparatus

In these experiments the Société[3] Genevoise spectrometer was used. The various adaptations of it to the different needs will be pointed out with reference to Fig. 1. X is the x-ray tube (a water-cooled molybdenum target tube of the Coolidge type), S_1, S_2, S_3 and S_4 are slits. C_1 and C_2 are calcite crystals, and P is the 90° calcite prism with the optic axis parallel to the 90° edge. P was mounted on a specially designed holder which permitted the

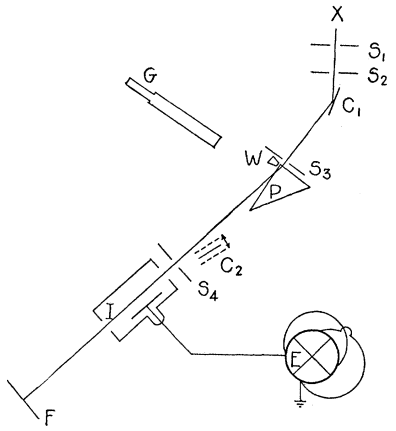

Fig.. 1

prism to be rotated about both horizontal and vertical axes in steps so small that any desired angular setting could be made easily. The holder also permitted the prism to be moved into and out of the beam laterally, keeping the faces of the prism accurately parallel to themselves. The 90° edge was in the axis of rotation of the prism mounting. The beam passed through this axis and nearly normal to one face of the prism. I is an ionization chamber and F is a photographic plate. The ionization currents were compared with a quadrant electrometer or a pliotron tube mounted over C_2. Methyl bromide (CH_3Br) vapor was used in the chamber to increase the absorption and ionization. The gain is due to the greater density of methyl bromide vapor as compared with air and to the higher K absorption of bromine for molybdenum rays. The angular positions of the prism were determined by means of an autocollimating telescope G, while the angular positions of C_1 and C_2 were determined by means of divided circles attached to the instrument. The divided circle attached to C_2 is presumably very good, being marked by the makers in 10 minute intervals which could be subdivided to 1 second by means of reading microscopes.

The slits, crystals and prisms were made parallel to the axis which supported C_2. This was accomplished by setting the axis of the autocollimating telescope perpendicular to the axis carrying C_2. To attain this a mirror with accurately parallel sides was placed on the table supporting C_2 and the autocollimating telescope and table were adjusted until the elevation of the beam reflected into the telescope did not change when the mirror was turned through 180°. The axis had previously been made vertical by means of a level. Since the telescope carried an accurate cathetometer level, the crystals on the mountings could be made parallel to the axis. Also by means of a steel mirror with accurately parallel sides placed between the jaws of the slits, the slits were made parallel to the axis. In the refraction experiments a lead wedge, W, was placed very close to the edge of the prism to stop as much as possible of the beam from getting by. The photographic plate was screened with lead from the direct beam practically all of the time during which a refraction photograph was being taken. This screen was removed during the last few minutes in order to obtain the direct beam from which to measure for the displacement of the refracted line.

To fix the angular position of the prism with respect to the beam, total reflection was used. P was rotated to different angular positions and the changes in intensity were noted by means of the electrometer readings. Having found the critical angle in this manner, the prism was turned into such a position that it would give total reflection. A photograph of the reflected line and the direct line on a plate at a known distance gave reliable data from which to compute the angle which the reflecting surface made with the beam. With this knowledge P could be turned to any desired angular position for refraction. The angles through which P was turned were measured by the autocollimating telescope, G, which had been calibrated previously against the circle attached to C_2. This telescope consisted of an object lens of 43 cm focal length and an eye combination which was the microscope of the Gaertner comparator used to measure the plates taken in this work. The microscope had a magnification of 28. When the reflector was turned through 1 second, the reflected image moved through 5.37 microscope divisions. The image reflected from the prism was such that the angles through which the prism was turned could easily be measured to 1 second.

Reflection and refraction were obtained for the heterogeneous radiation direct from the tube and for monochromatic radiation. When direct radiation was used, the arm carrying S_1, S_2 and C_1 was removed and the two slits were placed between P and the tube X. The double crystal arrangement permitted the determination of rocking curves and reflecting power of the crystals.

The measurement of the deviation of the refracted beam can be found in terms of the distance of the photographic plate from the refracting edge and the displacement on the plate of the refracted line from the undeviated line. Fig. 2 shows the rays and suggests how the measurements were taken.

The distance from F to A' was measured by pressing the end of a brass rod which had been squared in a lathe against the plate F and marking the point opposite A' with a sharp knife blade. This length was then measured

by comparison with a standard meter bar. The length AA' was measured with a traveling microscope. In the total reflection experiments the distance FA' was needed, while in the refraction experiments FA was needed. The edges of the lines measured to are clearly indicated in the diagram. Distances on the photographic plates were measured with a Gaertner traveling microscope reading to 0.001 mm. The calibration of the screw on the microscope was checked against a Zeiss scale and found to have no error larger than the errors of reading. That the plate (19 cm long) was satisfactorily perpendicular to the beam was checked by sighting along the arm of chamber I which was parallel to the beam.

The fact that the optic axis was parallel to the 90° edge of the prism was confirmed by a measurement of the indices of refraction of the ordinary and extraordinary rays on an optical spectrometer.

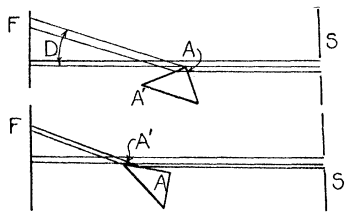

Fig. 2.

Experimental Results and Conclusions

As already mentioned, the specimen used in these experiments on total reflection and refraction was a 90° calcite prism having the optic axis parallel to the 90° edge. At first the two faces forming the 90° angle were highly polished. Later the polish on one face was removed by grinding in order to obtain a sharper edge, since the one left by the makers was a few hundredths of a millimeter thick. With the blunt edge it was impossible to obtain refraction at small angles of incidence on the emergent face. Total reflection from this edge was a disturbing feature. The dimensions of the faces forming the 90° angle were 10 mm×8 mm. A measurement of the ordinary and extraordinary indices of refraction for the Hg 5461 line gave 1.66169 and 1.4880 respectively as compared with the values 1.661647 and 1.487890 given by Baly[4] showing that the optic axis was very closely parallel to the 90° edge.

The experiments on total reflection, besides being useful in setting the prism for refraction, give some very interesting results. They give a determination of the index of refraction of calcite for the $K\alpha_1$ line of molybdenum as accurate at least as that obtained by the deviation method used by Davis-Hatley[1] and Hatley[2], who have determined the now accepted value. The broken curve, Fig. 3, shows the variation of the intensity of the reflected radiation in passing through the critical angle and the lack of any definite break shows there is no appreciable double refraction.

The prism was turned back to a point corresponding to a point on the

[4] E. C. C. Baly, Spectroscopy, Third Ed., Vol. 1, p. 83.

curve of maximum reflected intensity. A total reflection photograph gave 1.66×10^{-3} radians for the angle between the prism and the beam. Taking the mid-point of descent of the curve for the critical angle, we obtain for the critical angle 1.95×10^{-3} radians. The accuracy of this determination is limited in part by the choice of the critical angle from Fig. 3. Since the steep portion of the descent has an angular width of about 0.2×10^{-3} radians, the method has necessarily a possible error of 5 percent. Fig. 3 shows also in full line the theoretical curve for calcite. This was computed from the expression

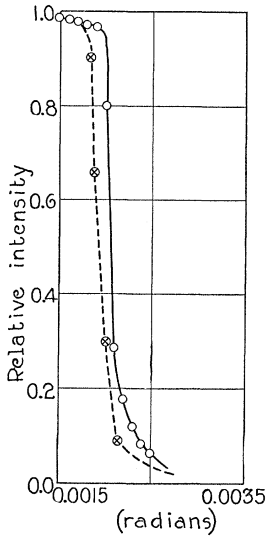

Fig. 3.

derived by Thibaud[5] for the ratio of the intensity of the reflected beam to that of the incident beam:

$$A = \frac{(1+m)^2 + 2(m^2+a^2)^{1/2} + 2(1+m)\left[2(m^2+a^2)^{1/2}\right]^{1/2}\cos\phi/2}{(1+m)^2 + 2(m^2+a^2)^{1/2} - 2(1+m)\left[2(m^2+a^2)^{1/2}\right]^{1/2}\cos\phi/2}$$

in which $\pi<\phi<2\pi$ and m is defined by the equation $\theta=(1+m)\theta_c$ where θ is the glancing angle of incidence and θ_c is the critical angle. The remaining quantities are defined in terms of the following relations:

$$K = k\lambda/4\pi;\quad a = K/2\delta;\quad \theta_c^2 = 2\delta;\quad \tan\phi = \left[-a(1-\delta)\right]/(m-a^2\delta);$$

$k\equiv$absorption coefficient; and $1-\delta\equiv$index of refraction. Table II gives the values of the constants used.

TABLE II.

k	20.6
a	2.91×10^{-3}
θ_c (radians)	2.00×10^{-3}
$\delta\times10^6$	2.00
$\lambda\times10^8$	0.709

[5] J. Thibaud, Jour. de Physique [7] **1**, 37 (1930).

k and δ were measured in these experiments, k being determined by the absorption in a plate of calcite 1 mm thick. The faces of the plate were parallel to cleavage planes. The curve shown is typical of several that were taken for monochromatic radiation. Curves were also taken for the direct radiation from the molybdenum tube. These had a descent somewhat broader than those obtained with monochromatic radiation. The tube, however, was operated at a voltage somewhat higher than the excitation voltage of the K-series of molybdenum. The voltage varied slightly in different experiments but was always approximately 40,000 peak. Total reflection was also obtained from a cleavage face of a calcite crystal at C_2. Here the descent of the intensity curve was very broad. A total reflection photograph from this cleavage face, which was a very good one as evidenced by its x-ray reflection of the Bragg type, gave a very broad and diffuse line. In fact it had several components. This crystal reflected 18.7 percent of the incident monochromatic radiation when crystals were in the antiparallel position. The tolerance angle was also measured for this position and was found to be 12 seconds in the first order.

The first experiments on refraction were performed with the direct radiation from the molybdenum target tube. The prism was set in the reflecting position by the process mentioned above. The prism was then turned to such a position that the angle ρ between the beam and the emerging face had the desired value, 9 minutes in the first experiment. A 5 hour exposure gave the refracted lines on a plate 166.6 cm from the refracting edge. The target of the tube was about 40 cm from this edge. ρ was made less in succeeding experiments until it was impossible to get refraction at 2 minutes. The edge was found not to be sharp. The edge was made sharp by grinding the entering face which had been highly polished. After the grinding it was not polished again. The refraction experiments were repeated, and it was found that good refracted lines could be obtained at values of ρ less than one minute. The lines were as sharp as they were before the entering face was ground, showing that the condition of the entering face has little part in refraction. The photographs showed a line due to the $K\alpha$ doublet and one due to the $K\beta$ doublet. Also there was radiation due to shorter wave-lengths. The fact that this refracted band of shorter wave-lengths came to a fairly sharp edge and did not reach the lines suggests that the short wave-lengths coming from the general radiation from the tube included considerable radiation from tungsten which had been deposited on the target. A study of the intensity curves,[6] due to A. W. Hull, for a molybdenum tube operating at the voltage used here (around 44,000 but not held constant) would not lead one to expect this edge if molybdenum radiation alone were present. On the other hand a comparison with the energy curve for a tungsten target[6] would lead one to expect this edge. A rough calculation (this could not be made precise because of the overexposure of the central line) confirmed the view that the band was due to general radiation.

[6] B. Davis, Phys. Rev. [2] **9**, 64 (1917).

Refraction was also obtained for an edge formed by two good cleavage faces from one of which total reflection was obtained as mentioned above. The refracted lines were very diffuse. Since the entering face does not effect refraction, then the emerging cleavage face is not suitable for refraction experiments. The fault could not be with the edge since the cleavage edge was much better than the edge on the 90° prism even after the grinding of the latter. These experiments on refraction with the direct radiation were not made very quantitative since the object was to get refraction with monochromatic radiation.

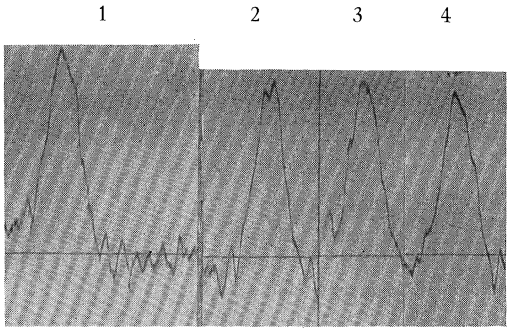

Fig. 4.

To obtain refraction with the $K\alpha_1$ line of molybdenum, C_1 was set to reflect $K\alpha_1$ at the best intensity. A good reflected line was obtained from C_1 in 2 seconds when the peak voltage was 45,000 volts and the current through the tube was 25 m.a. The angle ρ was made 7 minutes in the usual way. A good $K\alpha_1$ refracted line was obtained in 24 hours but on too dark a background to permit a close examination to be made of it. The wedge, W, was readjusted and ρ was made 1.45 minutes. The refracted line was good on a clear background. It was 0.077 mm broad and was free from a regular structure when examined under a microscope and by means of densitometer photographs (Fig. 4). Curves 1, 2, 3, and 4 were taken across different positions of this refracted line with a magnification of 40. Other refracted lines, less broad, obtained for smaller values of ρ and at longer exposures (up to 36 hours) also failed to show any fine structure which could be attributed to double refraction.

Fig. 5.

Fig. 5 shows the refracted line on the right and at the left the undeviated line from another of the good plates.

An attempt was made to find a possible fine structure by reflecting the refracted beam from C_2, but the beam was too weak to give an ionization cur-

rent measurable with an electrometer sensitivity such that the normal ionization did not mask the increase due to the refracted beam.

The refraction photographs permit a determination of the index of refraction of calcite for the $K\alpha_1$ line which has a smaller probable error than the now accepted value which was determined by the deviation from Bragg's formula. From the theory worked out by Stauss[7]

$$D + \rho = (\rho^2 + 2\delta)^{1/2}$$

in which D is the deviation due to refraction and δ is the dimensionless decrement of unity in the index of refraction. ρ has already been defined. In this equation the angles are to be expressed in radians.

Table III gives the data and results on the 11 plates taken. The distance FA' was 169.7 cm, AA' was 0.8 cm.

From the critical angle (1.95×10^{-3} radians) we get $\delta=1.90\times10^{-6}$. This value is subject to greater uncertainty than the others and is not averaged with them.

TABLE III.

Plate	d (mm)	ρ (radians)$\times10^3$	$\delta K\alpha_1\times10^6$
1.	3.096	0.192	1.993
2.	2.900	0.298	1.950
3.	2.743	0.432	1.941
4.	2.845	0.380	2.021
5.	2.838	0.380	2.012
6.	2.988	0.316	2.084
7.	2.227	0.920	2.050
8.	2.805	0.389	1.989
9.	2.313	0.791	1.989
10.	2.514	0.615	1.989
11.	2.840	0.360	1.995
		Mean	2.001 ± 0.009

It is the author's pleasure to acknowledge his indebtedness to Professor C. D. Cooksey for initiating this work, and to him, Professor L. W. McKeehan, and Mr. Donald Cooksey for much helpful advice throughout the course of the investigation. Also to Professor L. W. McKeehan the writer wishes to express his thanks for placing at his disposal the necessary facilities for doing the work.

[7] H. E. Stauss, J.O.S.A. **19**, 167 (1929).

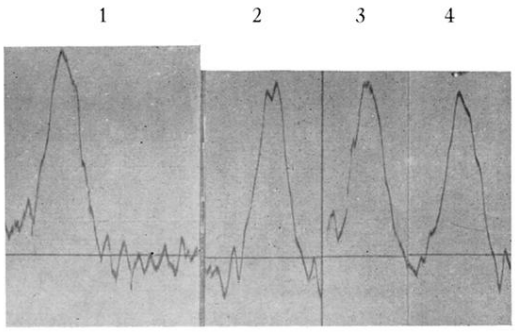

Fig. 4.

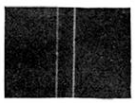

Fig. 5.